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Orthodontic resin containing bioactive glass: Preparation, physicochemical characterization, antimicrobial activity, bioactivity and bonding to enamel

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| ARTICLE INFO | A B S T R A C T |
|--|--|
| Keywords: Orthodontic composite Bioactive glass Bioactivity pH Ions release Antibacterial activity | <i>Objectives</i> : To synthesize experimental orthodontic resins used for bracket bonding containing different concentrations of niobophosphate (NbG) and 45S5 glasses. <i>Methods</i> : Experimental resins (Bis-GMA + UDMA + DMAEMA) were developed with two bioactive glasses (NbG and 45S5) in concentrations of 5, 10 and 20 (wt%). An experimental resin without glass and a commercial resin (Transbond XT) were used. Control of pH and ions release (calcium and phosphate) at different pH values (4 and 7) were evaluated in the time intervals of 24 h, 7 d, 14 d and 28 d. Microhardness, bioactivity (SEM, FTIR/ATR) and antibacterial activity of the resins were analyzed. Metal brackets were bonded to premolars (n = 10) with the resins to evaluate shear bond strength (SBS). <i>Results</i> : The experimental resins containing 45S5 were capable of raising the pH of the solution and showed high values of calcium and phosphate ions release. Resins containing NbG had a neutralizing potential. The NbG and Transbond resins released only phosphate ions. Transbond XT showed high microhardness values when compared with the experimental 45S5 resins ($p < 0.05$). Analysis by SEM showed precipitates and FTIR/ATR confirmed the presence of a calcium phosphate-based compound in the resins containing 45S5. There was no difference in SBS between the tested materials ($p > 0.05$). The resins with presence of bioactive particles showed antibacterial activity. <i>Conclusion</i> : Without compromising the shear bond strength, bioactive glasses showed the capacity to elevate pH, reduce the hardness of experimental resins when compared with Transbond XT and no filler experimental resin |

1. Introduction

One of the serious problems that may be caused by orthodontic treatment is the development of white spot lesions around brackets [1–3]. These lesions may develop in two ways: (1) due to phosphoric acid etching of a large area of enamel, without coating it with an adhesive system, which forms a rough surface that facilitates bacterial plaque accumulation [4]; (2) plaque accumulation due to the difficulty of cleaning around the bracket [5].

Previous studies have observed that white spot lesions may be

present in up to 97% of cases on conclusion of orthodontic treatment [6–8]. In an attempt to inhibit the appearance of these lesions, fluoride has been added to orthodontic resins to act on the process of enamel demineralization [9,10]. However, there is still no strong of evidence with respect to the effectiveness of including fluoride in bracket bonding materials [10,11].

At present, no material used for bracket bonding is capable of acting on pH control, antimicrobial activity and ion release, factors that may influence the process of enamel demineralization around brackets [12]. Thus, several researchers have added bioglass particles to various

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Table 1

Composition of experimental resin composites used.

| Material | Filler compo | osition (wt %) | Total filler composition (wt %) | Resin | | |
|------------------|--------------------|--------------------------------------|------------------------------------|---------|---|--|
| | Bioactive glass | Reinforcing fillers (Ba:Si – 2:1) | | wt % | Composition | |
| Transbond XT* | 0 | | | | Silane-treated quartz (70–80%), bisphenol A diglycidyl ether dimethacrylate (10–20%), bisphenol A bis(2-hydroxyethyl ether) dimethacrylate (5–10%), silane-treated silica (<2%) | |
| Control | 0 | 70 | 70 | 30 | 57.4% Bis-GMA | |
| 45S5/NbG | 5 | 65 | 70 | 30 | 42.6% UDMA | |
| 5% | | | | | Photoinitiator system: | |
| 45S5/NbG | 10 | 60 | 70 | 30 | 0.2% CQ | |
| 10% | | | | | 0.8% DMAEMA | |
| 45S5/NbG | 20 | 50 | 70 | 30 | | |
| 20% | | | | | | |

NbG: 40.1% Nb₂O₅, 32.8% P₂O₅, 21.2% CaO, Al₂O₃ 3.8%, 2.1% Na₂O, particle size d25 µm, silanization: none.

45S5: SiO₂ 45%, Na₂O 25%, CaO 25%, P₂O₅ 5%, particle size (d50/d99 μ m): 4.0/13.0, silanization: none, product name/manufacturer: G018-144/Schott, Germany. Barium-fillers (Ba): SiO₂ 55.0%, BaO 25.0%, B₂O₃ 10.0%, Al₂O₃ 10.0%, particle size (d50/d99 μ m): 1.0/4.0, silanization 3.2 wt%, product name/manufacturer: GM27884/Schott, Germany.

Silica-fillers (Si): SiO₂ ≥ 99.8%, primary particle size: 12 nm, silanization 4–6 wt%, product name/manufacturer: Aerosil DT/Evonik Degussa, Germany.

Bis-GMA: Bisphenol A glycidyl methacrylate, Sigma-Aldrich, USA; UDMA: Urethane dimethacrylate, Sigma-Aldrich, USA; CQ: camphorquinone, Sigma-Aldrich, WI, USA; 4E: ethyl-4- (dimethylamino) benzoate, Sigma-Aldrich.

materials for use in different fields of Dentistry [13–17]. On the other hand, the 45S5 glass has a high level of reactivity because of the release of a high quantity of (calcium and phosphate) ions that could interfere negatively in some of the properties of these materials.

More recently, some studies have suggested that the addition of niobium to the composition of bioactive glasses might increase the chemical stability of glass without interfering in the bioactivity of the material, due to its capacity to precipitate hydroxyapatite precursors [18–21]. Addition of bioactive niobophosphate glass (NbG) to different dental materials has shown promising results, such as: increase in radiopacity [22], ion-releasing capacity [23], bioactivity [22,24], pH control [25,26] and antibacterial activity [25]. In spite of these characteristics, up to now, no study has evaluated the influence of incorporating NbG into orthodontic resins on the properties of these materials.

Therefore, the aim of this study was to evaluate the pH, ion release, microhardness, antibacterial activity, bioactivity and bond strength of experimental resin materials containing different concentrations of NbG and 45S5 glass, and compare them with Transbond XT and an experimental conventional resin.

2. Materials and methods

2.1. Bioactive glasses

Two types of bioactive glasses were used: a commercial type (45S5, Sylc, OSspray Ltd, London, United Kingdom) and an experimental niobophosphate-based glass (NbG). The NbG and 45S5 glasses were crushed in a vibrating system (8000 M, Mixer/Mill, SPEX SamplePrep, NJ, USA) with tungsten carbide grinding balls-vial set (SPEX SamplePrep, NJ, USA) for 30 min. After grinding, the resultant powder was passed through a series of sieves of 150 μ m–75 μ m - 53 μ m–38 μ m - 20 μ m (Hogentogler & Co., Inc, Columbia, MD, USA). The particle size distribution was determined using a CILAS laser diffraction particle size analyzer (Model 1064, CILAS, Orléans, France). The particle size analysis showed a distribution of particles with a mean diameter of 5.92 μ m [26].

2.2. Preparation of experimental resins

Experimental resins containing different concentrations (5, 10 and 20 wt%) of two bioactive glasses (45S5 and NbG) were developed from the mixture of resin monomers and the glass particles. For the

experimental control material, a resin was developed with no bioactive glass addition (negative control group). The orthodontic resin Transbond XT (3 M Unitek, Monrovia, USA) was used as a commercial (positive) control group (Table 1).

All procedures were performed in a room equipped with a humidity ($50 \pm 5\%$) and temperature (23 ± 1 °C) controller (NOVOTEST TH802A, SP, Brazil). The monomers were weighed and mixed under magnetic stirring, and blended using a centrifugal mixing device (SpeedMixer, DAC 150.1 FVZ-K, Hauschild Engineering, Hamm, North Rhine-Westphalia, Germany). Thereafter, silica filler particles were added to the prepared monomer matrix and blended for 30 s at 3000 rpm, followed by bariumborosilicate glass filler for 1 min at 3500 rpm. Finally, the bioactive glass particles were added to the resins according to each experimental group and blended for 1 min at 3500 rpm. To eliminate possible porosities, each experimental composite was mixed one more time for 1 min at 3500 rpm under vacuum.

2.3. pH and ions release (calcium and phosphate)

Two solutions with different initial pH values (pH_i) were prepared. The pH_i 4 solution was prepared using hydrochloric acid (HCl) for adjustment, whereas, for the pH_i 7 solution, sodium hydroxide was used for adjustment. Discs of the tested materials were prepared with the use of a metal matrix (Ø 15 mm; thickness of 1 mm). The resins were inserted, covered with a polyester strip, pressed with a glass slide and light polymerized (Radii-cal, SDi, Victoria, Australia) for 20 s with a light intensity of 1200 mW/cm².

Four specimens of each group (n = 4) were made and individually immersed in plastic bottles containing 5 ml of each solution with pH_i and kept at a constant temperature of 37 °C. The pH was measured (QM-A338, Quimis, São Paulo, Brazil) in four different time intervals: 24 h, 7, 14 and 28 days. After each analysis, the specimens were removed from the jars, washed with deionized water, dried with absorbent paper and placed in new plastic bottles containing 5 ml of a fresh solution. Afterwards, the solutions were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES, ICP-9800, Shimadzu, Kyoto, Japan) to quantify the release of calcium and phosphate ions.

2.4. Knoop microhardness (KNH)

To evaluate the Knoop microhardness of each tested resin, four discs (n = 4) were fabricated in metal matrices (Ø 15 mm; thickness of 1 mm) in the same way as described in Item 2.3. The specimens were polished

with abrasive paper discs of different granulations (#600, #1200, #1600 and #2000) and felt discs with diamond particle ranging between 2 and 4 μ m (Diamond Excel, FGM, Joinville, Brazil). Subsequently the samples were stored in distilled water at 37 °C for 24 h and submitted to testing in a microhardness (HMV-G21DT, Shimadzu, Tokyo, Japan) with a load of 25 g for 5 s. The mean Knoop microhardness of each material was calculated from 4 indentations on the surface of each specimen, one in each quadrant.

2.5. Antibacterial activity

For antibacterial activity evaluation, six samples were manufactured in the same manner as described in item 2.3 and attached to the lid of a 24-well plate. The lid containing the samples was sterilized by UV light for 30 min. Aliquots of frozen stocks of Streptococcus mutans (UA159) were placed on Brain Heart Infusion (BHI; Sigma-Aldrich, St Louis, MO, USA) agar plates and incubated at 37 °C for 48 h. Colony-Forming Units (CFU) were collected and transferred to tubes containing BHI broth supplemented with 1% sucrose and grown until the late exponential phase [26,27]. In order to form a microbial inoculum, the suspensions were adjusted using the standard solution (0.5) according to the McFarland scale, resulting in a suspension with an approximate concentration of 10⁸ CFU/ml [28]. Aliquots of 1 ml of BHI supplemented with 1% sucrose were added to each well of a sterile 24-well plate followed by 100 µl of the adjustment microbial suspension. The lid containing the sterile samples was placed on the plate and incubated at 37 °C for 24 h. After 24 h, an aliquot of 100 µl from each well was transferred to tubes containing 1 ml of sterile 0.9% NaCl and vortexed vigorously. Aliquots of these suspensions were serially diluted up to 10^{-8} and 2 drops of 10 µl of each dilution were inoculated on BHI agar (BD, Sparks, USA) to determine the number of CFUs. The plates were incubated at 37 °C, 10% CO2 for 48 h, After 48 h, CFU were counted under stereomicroscope and the results were expressed as log₁₀ CFU/mL [27].

2.6. Bioactivity evaluation (SEM and FTIR/ATR)

Four discs from each group were made in the same way as described in Item 2.3. The specimens were stored in phosphate Buffered Saline Solution (PBS), (Dulbecco's Phosphate Buffered Saline, Sigma Adrich, St Louis, MO, USA) at 37 °C for 28 days. The specimens were taken to Scanning Electron Microscope (TM3030, Hitachi, Japan) for morphological analysis [22].

FTIR spectroscopy measurements were performed to identify characteristic mid-infrared bands of CaP (IRTracer-100, Shimadzu, Kyoto, Japan). Spectra were recorded in transmission mode between 2000 $\rm cm^{-1}$ and 400 $\rm cm^{-1}$, with a resolution of 4 $\rm cm^{-1}$, using KBr pellets.

2.7. Shear bond strength test (SBS)

The sample size was estimated based on the number of brackets required because this was the unit of measurement. A sample size of 80 brackets (n = 10) was sufficient to detect a difference with 80% power and a 5% significance level (G*Power 3.0.10, Franz Faul, Universität Kiel, Germany). A total of 80 human maxillary premolars free of caries, cracks, and restorations were used. These teeth had been extracted for orthodontic reasons and were used with the informed consent of the patients. Ethical approval for collection of the teeth was obtained from the Local Federal University Committee (2.496.044). The teeth were washed in water and stored in a 0.1% thymol solution, for no longer than 2–3 months before use.

After this, the specimens were randomly divided into 8 groups and their vestibular surfaces were etched with 37% phosphoric acid (Condac 37, FGM, Santa Catarina, Brazil) for 30 s. After being washed and dried, a thin coat of adhesive Primer Transbond XT (3 M Unitek, Monrovia, USA) was applied. Light polymerization was performed with an LED Radii-cal (SDI, Victoria, Australia) light appliance, and Standard Edgewise (3 M Unitek, Monrovia, USA) metal brackets for premolars were bonded to the specimens with the resin of each experimental group. In the same manner, each proximal surface was light activated for 20s.

After bracket bonding, the teeth were embedded in acrylic resin, in vertical position, to perform the SBS test. To ensure that the force of the chisel would fall perpendicularly on the tooth/adhesive interface, the teeth were positioned with the help of a prosthetic delineator. The specimens were taken to the Universal Test machine (Instron 3342, Canton, USA) with the force applied in the occlusal-vestibular direction at a speed of 1.0 mm/min (Odeme Biotechnology, Luzerna, Santa Catarina, Brazil). The shear force used to debond each bracket was recorded in Newtons (N) and converted into Mega-Pascal (MPa), as the ratio of Newton on the surface area of each bracket (MPa = N/mm²).

After debonding, the teeth and brackets were examined at $10 \times$ magnification under a stereoscopic magnifying glass (Kozo Optical and Electronical Instrument Co, Nanking, China) by a blinded operator, using the adhesive remnant index (ARI) to describe the quantity of resin remaining on the tooth surfaces [29]. The ARI scores (ranging between 0 and 3) obtained were as follows: 0, no adhesive remained on the tooth; 1, less than half of the enamel bonding site was covered with adhesive; and 3, the enamel bonding site was covered entirely with adhesive.

2.8. Statistical analysis

For statistical analysis, the SigmaPlot software (Systat Software Inc., San Jose, California, USA) was used. After confirming normal distribution of the sample by means of the Shapiro-Wilk test ($\alpha = 0.05$), the KNH and SBS data were submitted to one-way analysis of variance (One-Way ANOVA) and the Holm-Sidak test for contrast between the means ($\alpha = 0.05$). To compare the antimicrobial activity, the colony forming unit (CFU) data were transformed into Log 10 (CFU/mL) and submitted to the Kruskal-Wallis and Student-Newman-Keuls post-hoc test. The pH, ion release and FTIR/SEM analysis were shown in descriptive values.

3. Results

3.1. pH and ion release analysis

During all time intervals determined for pH analysis, the resins containing 45S5 (5, 10 and 20%) were capable of alkalize the solutions. The resins containing NbG glass had the capacity to neutralize the solutions, irrespective of the initial pH. Transbond XT and the resin without bioactive glass showed a pH below 7 in the measured time intervals (Fig. 1).

The ICP-AES analysis showed that the materials Transbond XT, experimental control (no filler) and the resins containing NbG particles were incapable of releasing calcium ions. Whereas, the resins that contained 45S5 glass released a large volume of calcium ions in both (4 and 7 pH) solutions. The group containing 20% of 45S5 glass released the highest volume of calcium ions (Fig. 2A).

Phosphate ions release was observed in almost all groups, with the exception of the experimental control (no filler). The highest volume of phosphate ions release was found in the groups containing particles of 45S5 glass, irrespective of pH. A low volume of phosphate ion release was observed in the groups of resin containing NbG glass and Transbond XT (Fig. 2B).

3.2. Knoop microhardness (KNH)

The Knoop microhardness values of each orthodontic resin are presented in Fig. 3. For the groups containing 45S5, the KNH measurements were significantly lower than those of the Transbond XT (p < 0.05). The 45S5 20% group had a lower KNH value than those of the experimental control (no-filler) and NbG 5% groups (p < 0.05). The Transbond XT,



Fig. 1. pH changes over time for both glasses in different concentrations: (A) initial pH 4.0 and (B) initial pH 7.0.



Fig. 2. Results of ions release of resin orthodontics: (A) calcium ion release (B) phosphate ion release. Each value is the mean of three measurements, with the error bar showing standard deviation (mean \pm sd; n = 4).

experimental control (no filler) and resin containing NbG glass in the different concentrations showed similar Knoop microhardness values (p > 0.05).

Streptococcus mutans, when compared with the Transbond XT and experimental control (no filler) resins. The Transbond XT and control (no filler) resins had similar antibacterial activity values.

3.3. Antibacterial activity

Fig. 4 shows the results of antibacterial activity evaluation of the tested materials. The experimental resins containing different concentrations of NbG and 45S5 glasses showed a reduction in CFUs of

3.4. Bioactivity (SEM – FITR/ATR)

In the images of the control (no filler) and Transbond XT groups, no formation of precipitates was observed on the surface of these materials (Fig. 5). Whereas, the resins that contained bioactive glasses showed



Fig. 3. Knoop Microhardness of resin orthodontics tested: Each value is the mean of four measurements, with the error bar showing one standard deviation (mean \pm sd; n = 4). Bars with dissimilar letters indicate values that are significantly different from each other (p < 0.05).



Fig. 4. Results of antibacterial activity of resin orthodontics are expressed as mean \log_{10} CFU. Box-plot with different letters indicates values that are significantly different from each other (p < 0.05).

formations with the appearance of spherical agglomerates. Furthermore, the formation of a large volume of these crystals was observed in the resins containing 45S5. In the resins containing NbG, it was possible to observe discrete deposition of precipitates.

FTIR/ATR analysis showed peaks of 1418 and 1488 cm⁻¹ (calcium

carbonate) and 710 and 640 cm⁻¹ (phosphate) on specimen surfaces of 45S5. NbG specimens showed only phosphate peaks 640 cm⁻¹ (Fig. 6). Transbond XT and experimental control resin showed no peaks of hydroxyapatite precursors.



Fig. 5. Scanning electron microscopy of the tested materials after 28 days immense in PBS (Magnification $5000 \times -$ Bar 20 µm): (A) Control resin (no fillers): It is possible to observe the presence of fillers particles embedded in the resin matrix and the absence of precipitate formation (B) Resin 45S5 5%: Formation of aggregates in a small quantity on the surface of the resinous material (white arrow). (C) Resin 45S5 10%: Spheroidal precipitates were deposited on the surface to form clusters throughout the resin surface (white arrow). (D) Resin 45S5 20%: Spheroidal precipitates were deposited on the surface and packed together to form clusters of aggregated spherulites evenly distributed throughout the cement surface (white arrow). (E) Transbond XT: Presence of fillers particles and the absence of precipitate formation. (F) Resin NbG 5%: It is possible to observe a large bioactive NbG glass particle and on its surface the presence of spherical precipitates (white arrow). (G) Resin NbG 10%: Presence of precipitates in a small quantity on the surface of the material (white arrow). (H) Resin NbG 20%: Presence of few clusters on the surface of the resin (white arrow).



Fig. 6. FTIR/ATR spectrum of materials tested after 28 days immersed in PBS (A) 45S5 groups indicate the presence of phosphate: 640 cm^{-1} , 710 cm^{-1} ; and carbonate groups: 1418 cm^{-1} , 1488 cm^{-1} (B) NbG groups show only presence peaks of phosphate: 640 cm^{-1} .

| Table 2 |
|--|
| Shear bond strength values and adhesive remnant index (ARI) scores for the |
| orthodontic resins materials tested in the present study. |

| Groups | Mean (MPa) | ARI | | | | |
|--------------|-----------------------------|-----|---|---|---|--|
| | | 0 | 1 | 2 | 3 | |
| Transbond XT | $11.7\pm1.9^{a,b,c}$ | 3 | 3 | 1 | 3 | |
| Control | $8.9\pm1.6^{\rm c}$ | 8 | 1 | 1 | 0 | |
| NbG 5% | $11.0\pm1.6^{\rm b,c}$ | 8 | 1 | 1 | 0 | |
| NbG 10% | $13.0\pm2.4^{\rm a,b}$ | 4 | 4 | 0 | 1 | |
| NbG 20% | $13.9\pm1.5^{\rm a}$ | 4 | 2 | 2 | 1 | |
| 45S5 5% | $11.4\pm2.4^{\rm a,c}$ | 4 | 2 | 1 | 1 | |
| 45S5 10% | $13.3\pm1.9^{\mathrm{a,b}}$ | 5 | 2 | 1 | 1 | |
| 45S5 20% | $12.4\pm1.8^{\rm a,b}$ | 2 | 6 | 0 | 1 | |

Different superscripts indicate significantly different value (p < 0.05).

3.5. Shear bond strength test

The means and standard deviations of the shear bond strength test and description of the adhesive remnant index are presented in Table 2. One-Way ANOVA demonstrated that there was difference in the bond strength values between the experimental control resin (no filler) and groups with NbG 10%, NbG 20%, 45S5 10% and 45S5 20% glasses (p < 0.05). The bond strength value of Transbond XT was similar to those of all the other tested resins (p > 0.05).

The fracture mode results of the tested experimental materials had a similar behavior, in which it was possible to observe an ARI value of between 0 and 1. Transbond showed the highest frequency of ARI = 3 (Table 2).

4. Discussion

The discovery of a material with the ability to reduce the changes suffered by enamel in the bracket bonding procedures may be decisive for preventing the sequelae of orthodontic treatment. Properties such as antibacterial action [14,27,30,31], pH control, maintenance of the mechanical properties of enamel [32], inhibition of demineralization [13, 33–35] and potential for remineralization are ideal for this material [15, 27,36,37]. The present study showed promising results for the experimental resins containing bioactive glasses, when compared with an experimental control resin (no filler) and a gold standard resin found on the market (Transbond XT).

In the pH results, it was possible to observe that the resins containing 45S5 had a strong potential to alkalize the medium, irrespective of the initial pH (4 or 7) (Fig. 1). The alkaline pH of bioglass 45S5 was due to a rapid release of Na⁺ or K⁺ and the incorporation of H₊ or H₃O⁺ into the glass particles [16,38]. This alkaline pH produced by resins containing 45S5 glass appeared to guide the behavior of the material in ion release, antibacterial activity and in precipitate formation [38,39].

In this study, a large volume of precipitates formed on the surfaces of the resins containing 45S5 was observed, irrespective of the concentration of glass (Fig. 5B, C, D). The most appropriate pH to favor the formation of stoichiometric nano-hydroxyapatite ranges between 8 and 10 [40]. These precipitates are presented in the form of clusters of spherical particles, probably a deposit of Ca–P [41]. The FTIR/ATR analysis confirmed the presence of apatite precursor compounds, in which it was possible to observe peaks of calcium carbonate (1488 cm⁻¹ and 1418 cm⁻¹) and phosphate (710 and 650 cm⁻¹).

Previous studies have confirmed the high capacity of 45S5 glass to alkalize pH [39,42,43]. This increase in pH also plays an important role in antibacterial activity [44]. The antibacterial effect of 45S5 may have included two aspects: (1) the higher pH proximal to the 45S5 particles, which were closely surrounded by bacteria; (2) the destruction of the cell walls by 45S5 debris [44,45]. In addition, we believe that 45S5 and NbG precipitates or debris could damage cell walls and lead to bacterial death. But the mechanisms of the antibacterial effect of the two bioactive glasses were still under discussion.

In the present study it was also possible to observe the capacity of NbG glass to neutralize the pH, irrespective of the initial condition (Fig. 1). On the other hand, NbG glass released only phosphate ions, and no calcium ion release was observed. Previous studies have demonstrated the capacity of this glass to neutralize pH and release ions (calcium and phosphate) [26], but in lower quantities when compared with 45S5 [25]. The fact of not having found calcium ions released in the solution could be because they were trapped in the polymeric matrix of the experimental resin, which prevented the ions from leaving in large volumes. This was more evident because previous studies performed the readout of ion release only in the NbG glass powder [25].

The low volume of ion release of the materials containing NbG glass in this study reflected on the formation of few precipitates on the surfaces of NbG glass specimens, on which only peaks of phosphate precipitates were found (Fig. 6B). However, peaks of calcium and phosphorous have previously been found in materials doped with a high volume (30% wt) of NbG glass [22]. Even with the lower results of pH and ion release, the compounds containing NbG glass showed good results for antibacterial activity, which have also been found in previous studies [25,26]. Zehnder et al., 2004 [46] reported that S53P4 bioactive glass powder had a greater antiseptic effect than that observed for calcium hydroxide and that apparently, this effect was not related to pH alone. Carneiro et al., 2018 [22] found precipitates on the surface of a material doped with NbG glass; this in conjunction with the release of other elements such a sodium and niobium may explain the antibacterial effect of this material.

Transbond XT resin presented results that ranged from 4.6 to 6.5 in the tested pH solutions. These results, found in conjunction with no release of calcium and phosphate ions and absence of precipitates, may be the explanation for the low performance of this material in the antibacterial activity test. The results obtained for both Transbond XT and experimental control (no filler) resins may be the explanation for the high rates of white spot lesions found at the end of orthodontic treatments [6].

Some studies have shown that in the presence of sucrose, the oral plaque pH can decrease to 4.5 or even 4. A plaque pH of higher than 6 is considered to be in a safe range, a plaque pH of 6.0–5.5 are in the potentially cariogenic range, and pH of 5.5–4 are the cariogenic or danger range for cavity formation [47–49]. Xu et al., 2009 [49] showed that it is fundamental for composites to be "smart": to increase the release of caries-inhibiting ions at a lower pH, when the calcium and phosphate ions are most needed. This ion release triggered by a local drop in pH may help to prevent demineralization in tooth structures contiguous to the "smart" composite restoration.

Another desirable characteristic for orthodontic resins is a low wear resistance, which facilitates their removal after conclusion of orthodontic treatment. Iijima et al., 2008 [50] conducted a study to evaluate the grindability of orthodontic resins by evaluating parameters such as hardness of the material. They observed that materials containing high concentrations of SiO₂, Ba₂O₃ and Al₂O₃ such as Transbond XT resin have high resistance to wear. Perhaps this is why Transbond XT resin showed higher hardness values than all the experimental resins containing 45S5 glass.

The high level of chemical reactivity of 45S5 glass in an aqueous medium may be related to its high ion release capacity, high potential to raise the pH and antibacterial activity. This process appears to culminate in structural loss due to dissolution, which could reduce its microhardness. This may be related to the fact that the resin with 20% of 45S5 glass showed the highest capacity to raise the pH of all the solutions, but also was the resin that had the lowest hardness values.

Transbond XT resin had a higher ARI value in comparison with those of the other materials, with 70% of the teeth in this group requiring mechanical removal of the adhesive remnant. This high wear resistance may demand a longer chair time spent by the professional, in addition to a possibility of damage to the enamel due to the use of burs and discs [51,52].

The addition of bioactive glasses to orthodontic resins favored the obtainment of excellent results, and this did not interfered negatively in the bond strength values of these materials, irrespective of concentration and bioactive glasses used; the results obtained by the experimental resins were statistically similar to those of Transbond XT resin.

High bond strength values between the tooth and bracket have always been an important factor in studies for developing a material for orthodontic bonding. However, the development of a "smart material" resin, capable of controlling the pH, inhibiting bacterial action and releasing ions that enable the formation of precipitates that can remineralize the tissues around orthodontic brackets, may become a new trend for the future of orthodontic materials.

5. Conclusion

The bioactive glasses showed the capacity to elevate pH, reduce the hardness of experimental resins when compared with Transbond XT and an experimental resin (no filler). Furthermore, only resins containing bioactive glasses presented the formation of structures similar to crystals on their surfaces, which could be an indication of the bioactivity of these materials. All of these characteristics were obtained without compromising the bond strength values.

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